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Effect of substrate temperature on microstructure and optical properties of nanocrystalline alumina thin films

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Abstract

Aluminum oxide (Al_2O_3) thin films were deposited on silicon (100) and quartz substrates by pulsed laser deposition (PLD) at an optimized oxygen partial pressure of 3.0×10^{-3} mbar in the substrate temperatures range 300–973 K. The films were characterized by X-ray diffraction, transmission electron microscopy, atomic force microscopy, spectroscopic ellipsometry, UV–visible spectroscopy and nanoindentation. The X-ray diffraction studies showed that the films deposited at low substrate temperatures (300–673 K) were amorphous Al_2O_3, whereas those deposited at higher temperatures (≥773 K) were polycrystalline cubic γ-Al_2O_3. The transmission electron microscopy studies of the film prepared at 673 K, showed diffuse ring pattern indicating the amorphous nature of Al_2O_3. The surface morphology of the films was examined by atomic force microscopy showing dense and uniform nanostructures with increased surface roughness from 0.3 to 2.3 nm with increasing substrate temperature. The optical studies were carried out by ellipsometry in the energy range 1.5–5.5 eV and revealed that the refractive index increased from 1.69 to 1.75 ($\lambda=632.8$ nm) with increasing substrate temperature. The UV–visible spectroscopy analysis indicated higher transmittance (> 80%) for all the films. Nanoindentation studies revealed the hardness values of 20.8 and 24.7 GPa for the films prepared at 300 K and 973 K respectively.

Keywords: C. Optical properties; Thin films; Alumina; Pulsed laser deposition; X-ray diffraction

1. Introduction

Aluminum oxide (Al_2O_3) thin films possess the excellent properties such as high melting point, high hardness, medium refractive index, high transparency, low absorption, wide bandgap, high thermal conductivity, low electrical conductivity, high radiation resistance, high corrosion resistance with good chemical and thermal stability [1–3]. Hence, the Al_2O_3 thin films are used as buffer layer for silicon-on insulator devices, gate insulator for metal-oxide-semiconductor devices, metal-nitride-oxide-semiconductor and complementary metal-oxide-semiconductor devices [4–8]. Al_2O_3 thin films are used in anti-reflection coatings, water repellent coatings, organic light emitting devices and improving the adhesion of bioactive glasses and hydroxyapatite coatings for medical implants [9–14]. They also find many important applications in opto-electronics, wear and corrosion resistant coatings for cutting tools, oxygen permeation barrier coatings for turbine blades, hydrogen permeation barrier coatings for nuclear fusion reactors, bioceramic, catalysis, and sensors [15–18]. The surface properties of various materials such as glasses, metals, bioceramics and polymers can be modified using Al_2O_3

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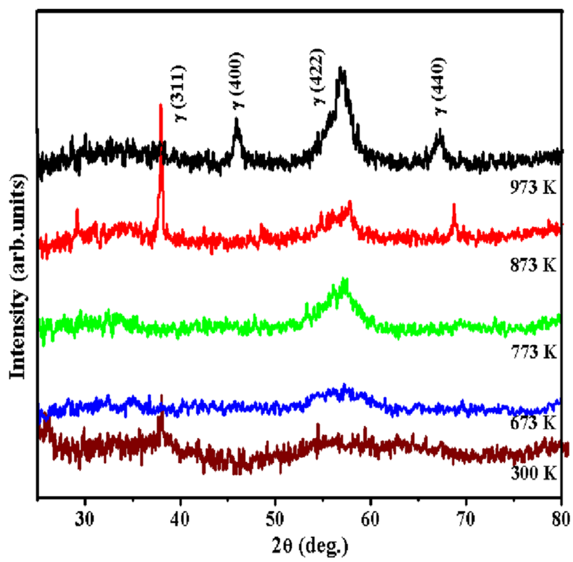

Fig. 1. XRD pattern of the Al_2O_3 films prepared at various substrate temperatures at an oxygen partial pressure of 3×10^{-3} mbar.

coatings [19]. The anti-reflection coatings are commonly used in instrument panels, displays, camera lenses, binoculars and telescopes to help produce clear and sharper images and all these applications require smooth films with stoichiometry and low defects density. It is well known that the properties are dependent on the preparation method and processing parameters. Hence it is important to investigate the correlation among the process parameters, microstructure and properties for several technological applications.

Al_2O_3 exhibits different crystallographic polymorphs such as γ, η, θ, δ and α phases with respect to temperature. Among the different phases of Al_2O_3, γ phase occurs in low temperature and find several technological applications [20]. Al_2O_3 thin films are deposited by different methods [21–25]. Among these methods, the pulsed laser deposition (PLD) is very flexible, simple, free from contamination, fast and controllable method for making high quality thin films of metals, semiconductors, insulators, polymers and biological materials. There are several reports on the preparation of Al_2O_3 thin films by PLD. However, there is no systematic study on microstructural, optical and mechanical properties of γ-Al_2O_3 films prepared as a function of substrate temperature. Hence, the present investigation deals with the preparation of Al_2O_3 thin films on Si (100) and quartz substrates as a function of substrate temperature (300–973 K) in order to understand the influence of substrate temperature on the microstructure, optical and mechanical properties of the films.

2. Experimental details

Al_2O_3 (99.99% purity) powder was compacted into a pellet of 25 mm diameter and 3 mm thickness at a pressure of 10 MPa using a uni-axial press. The pellet was sintered at 1473 K for 6 h. PLD experiments were performed using a KrF excimer laser (λ=248 nm) and the other deposition parameters are given elsewhere [23]. The thickness of the films was measured by Dektak profilometer (DEKTAK 6M-stylus profiler). The crystallinity of films were studied in an INEL XRG-3000 X-ray diffractometer (GIXRD) using CuKα_1 radiation. HRTEM investigations were carried out on a JEOL 2000 EX II (*T*) operated at 200 kV. The surface morphology and root mean square (RMS) surface roughness of the films were examined by Nanoscope E (Digital instruments Inc., Model: NSE, USA) atomic force microscope (AFM) in contact mode. The optical properties were measured by a SOPRA ESVG model rotating polarizer ellipsometer in the energy range 1.5–5.5 eV for three different angles of incidence (65°, 70° and 75°). The optical properties were also measured by a UV–vis–NIR (model no:3101/PC, Shimadzu) spectrophotometer in the wavelength range 190–800 nm. The mechanical measurements were carried out by nanoindenter (CSM, Switzerland) equipped with a Berkovich diamond indenter tip.

3. Results and discussion

3.1. Microstructural characterization

3.1.1. XRD, TEM and AFM analysis

The sintered Al_2O_3 pellet (α-Al_2O_3 of hexagonal structure, a=4.75 Å, and c=12.99 Å) was used as a target for PLD [23]. Al_2O_3 thin films were deposited on Si (100) and quartz substrates by PLD in the substrate temperatures range 300–973 K. Fig. 1 shows the XRD pattern of the Al_2O_3 films and revealed that the films were amorphous at low substrate temperatures (300–673 K), and polycrystalline Al_2O_3 in the temperatures ≥773 K. The films deposited in the temperatures ≥773 K indicated (311), (400), (422) and (440) reflections, correspond to γ-Al_2O_3 of face centered cubic structure [26,27]. The peak intensities of the films increased with increasing substrate temperatures. The mean crystallite size was determined from the Scherrer formula after subtracting the instrumental broadening. The crystallite size was found to increase from 5 to 10 nm as the temperature increased from 300 K to 973 K. The phase formation depends on the preparation method and process parameters. In general, phase formation of Al_2O_3 depends strongly on the substrate temperature [28]. It was reported by Cibert et al. [24] that the films prepared by plasma enhanced chemical vapor deposition (PECVD) at room temperature were amorphous, whereas the films showed cubic γ-Al_2O_3 structure at 1073 K. Also, the γ-Al_2O_3 films prepared by the PLD technique were reported to be formed at a substrate temperature of 1063 K only [29]. Pradhan et al. [30] have deposited Al_2O_3 thin films by metal organic chemical vapor deposition as a function of temperature. The films were amorphous at the low temperature range 623–823 K and crystalline Al_2O_3 phase in the temperature range 823–1023 K. Zywitzki et al. [22] have reported the nanocrystalline γ-Al_2O_3 layers with a grain size of 12–15 nm at a substrate temperature of 973 K. Anders et al. [31] reported the α-phase Al_2O_3 by post-annealing at 1273 K for 16 h. In the present work, γ-Al_2O_3 films were obtained at a low substrate

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